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1 *In situ* granulation by thermal stress during subaqueous
2 volcanic eruptions

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15 **ABSTRACT**

16 Some of the most complex volcanic thermodynamic processes occur when
17 erupting magma interacts with water. In shallow water, “Surtseyan” eruptions are
18 spectacular, and they efficiently fragment magma into fine ash particles. The aviation
19 hazard from these eruptions depends the amount of transportable fine ash that is
20 generated and whether it is aggregated into particle coatings or accretions. To investigate
21 both mechanisms, we analyzed ash-encased lapilli from the Surtseyan eruptions of
22 Capelinhos (Azores, 1957–1958) and Hunga Tonga–Hunga Ha’apai (Tonga, 2014–2015)

using X-ray computed microtomography and electron microscopy. We discovered pyroclasts that were not coated, *sensu stricto*, but had enveloping ash produced by *in situ* granulation of the particle surface. This **was** caused by thermal stress as pyroclasts briefly traveled through water and were quenched during eruption. *In situ* granulation is thus an important secondary disruption process in shallow subaqueous eruptions. Our results imply that ash encasement is not always evidence of particle aggregation and accretion, but it may also result from new ash formation. Shallow-water conditions produce the most efficient ash-generation conditions, leading to the greatest hazard to downwind populations and air traffic.

INTRODUCTION

Surtseyan eruptions are explosive volcanic events characterized by spectacular explosive jets that burst through ocean or lake water. They were named after the A.D. 1963–1967 submarine eruption of Surtsey, Iceland (Thorarinsson, 1967), which followed a similar well-observed eruption at Capelinhos in the Azores (1957–1958). One of the most recent eruptions of this type was at Hunga Tonga–Hunga Ha’apai (**Tonga**) in 2014–2015, which caused cancellation of international flights due to widespread ash dispersal (Global Volcanism Program, <https://volcano.si.edu/>; Colombier et al., 2018; Garvin et al., 2018). Magma interaction with shallow groundwater or surface water often produces copious volumes of fine volcanic ash, which is particularly hazardous to air transport, such as the subglacial Eyafjallajökull event of 2010 (e.g., Gudmundsson et al., 2012). Water is a far more efficient cooling medium than air, so that shock magma quenching is a primary fragmentation mechanism (Zimanowski et al., 1997), alongside gas-driven magmatic fragmentation (Gonnermann, 2015). Water may also drive secondary magma

disruption via steam-driven explosions (Kilgour et al., 2010), thermal granulation (Kokelaar, 1986), and turbulent shedding (Mastin, 2007). The rapid cooling of fragmenting magma also influences eruption dynamics by hindering vesiculation and causing particle aggregation and recycling (Cole et al., 2001; Schipper and White, 2016). Moore (1985) noted that ash-“coated” lapilli were the dominant constituent of the deposits at Surtsey, and aggregation of ash particles was subsequently recognized as a common process during magma-water interaction (e.g., Brown et al., 2012).

Aggregation of particles as accretionary lapilli or pyroclast coatings may lead to a lower-than-expected ash dispersal and hazard. To examine the formation and stability of ash haloes, we used three-dimensional (3-D) X-ray computed microtomography (micro-XCT) on particles from Surtseyan eruptions (Capelinhos 1957–1958 and Hunga Tonga–Hunga Ha’apai 2015–2016). We aimed to quantify the effectiveness of the aggregation processes implied in the formation of these particle types (e.g., Mueller, 2013). Instead, we discovered that these reflect additional post-eruptive disruption via thermal granulation.

METHODS

Using micro-XCT, we investigated four ash-encased lapilli from the basaltic andesitic to andesitic eruption of Hunga Tonga–Hunga Ha’apai volcano (e.g., Colombier et al., 2018) and three ash-encased lapilli from the basaltic eruption of Capelinhos (Zanon et al., 2013; for a detailed description of the eruption deposits and analytical conditions, see the GSA Data Repository¹). Two bombs from the magmatic phase of the Capelinhos eruption were analyzed for thermal expansivity and determination of the glass transition temperature. We measured the thermal expansivity using a NETZSCH Dilatometer 402C

with a heating rate of 10 K min^{-1} in argon. The linear expansion coefficient measured between room temperature and $T = 550 \text{ }^{\circ}\text{C}$ was $\alpha = 5.5 \times 10^{-6} \text{ K}^{-1}$. A differential scanning calorimeter (DSC) 404C calibrated for temperature and sensitivity was used, and the DSC signal was acquired during heating at 10 K min^{-1} in argon. The glass transition temperature determined by both expansivity and DSC signal was approximately $T_g \sim 600 \text{ }^{\circ}\text{C}$.

NEW OBSERVATIONS FROM MICROTOMOGRAPHY

Ash-encased lapilli are the dominant constituent (>90%) of the 2–5 mm particle size in fall and surge deposits from Capelinhos and Hunga Tonga–Hunga Ha’apai. Their cores consist of scoria with variable vesicularity, vesicle-size distribution, vesicle shape, and crystallinity (Figs. 1A–1C). They are partially to completely surrounded by a $\leq 500\text{-}\mu\text{m}$ -thick rim of fine ash particles (Figs. 1A–1C), frequently filling the external vesicles at the clast margin (Figs. 1G–1I and 2D). The interface between scoria cores and the ash rim is characterized by abundant cracks, sometimes organized in clusters (Figs. 1D–1I). Particles were not cut or mechanically processed, so the fractures are natural. In partially ash-encased lapilli, fractures occur only where ash rims exist (Figs. 1C, 1F, and 1I). The cracks extend into the scoria and are commonly planar to curvilinear (Fig. 1I).

In the ash rim, and at the contact to the core, we observed matching jigsaw-fit fracture planes (Figs. 1H, 2A, and 2B), vesicle concavities (Figs. 1H and 2C), and crystal fragments (Figs. 2A and 2B). Many of the jigsaw-textured domains showed varying degrees of internal particle rotation and/or displacement down to the scale of the smallest particles ($\sim 20 \text{ }\mu\text{m}$) clearly observable with the micro-XCT resolution. The crack number density decreased from the ash rim toward the scoria core (Figs. 1 and 2).

We found that the heterogeneous vesicle and crystal textures strongly influenced the number, density, size, and geometry of the cracks in the particles observed. Vesicles with regular crack spacing demonstrated radial stress concentration and crack activation (Heap et al., 2014; Figs. 2D and 2E). Cracks also commonly diverted at interfaces between phases, showing propagation along crystal-glass boundaries (Fig. DR1 in the Data Repository). These features influenced crack number, density, and size as well as the grain size and morphology of resulting ash particles, similar to that reported elsewhere (Liu et al., 2015).

ORIGIN OF THE ASH-ENCASED LAPILLI

The jigsaw fit of the ash rims to the host particles indicates that they formed by brittle granulation of the particle margins. That is, these are not “coated” particles, *sensu stricto*. The clear shape correspondence and packing of neighboring particles in three dimensions show that that they were formed *in situ* by disruption of the porous scoria. The outward increase in the crack number and density from the core to the ash rims implies a continuous transition between cracking and granulation. Therefore, the cracks and jigsaw-fit textures were formed by the same brittle process.

The presence of jigsaw textures in subaqueous settings has been reported in studies of hyaloclastites and peperites (Carlisle, 1963; Staudigel and Schmincke, 1984; Hanson and Hargrove, 1999; Doyle, 2000; Skilling et al., 2002) and is generally attributed to *in situ* thermal granulation during water-magma interaction. In this process, and in contrast to explosive fragmentation mechanisms (cf. Carlisle, 1963; van Otterloo et al., 2015), particles remain at their sites of formation. By analogy, we propose that

after explosive magma fragmentation, the margins of the lapilli-sized clasts examined here experienced thermal cracking and granulation due to quenching in seawater.

During quenching, the margins of particles experience the highest local cooling rates and thermal stress. In the case of direct contact with water, where vapor films are absent or collapse, we can assume that the surface of the particles is instantaneously cooled to the water temperature ($T_w \sim 25^\circ\text{C}$). Therefore, instant thermal stress σ can then be calculated using (van Otterloo et al., 2015):

$$\sigma = (E\alpha\Delta T)/(1 - \nu) , \quad (1)$$

where E is the elastic modulus ($E = 73\text{ GPa}$; Schultz, 1993), α is the thermal expansivity ($\alpha = 5.5 \times 10^{-6}\text{ K}^{-1}$), ν is Poisson's ratio ($\nu = 0.25$; Schultz, 1993), and ΔT is the quenching temperature difference. For the initial melt temperature, we chose a minimum value that corresponds to the glass transition temperature T_g (600°C). The temperature difference ΔT is therefore $T_g - T_w = 575^\circ\text{C}$. This yields an instant thermal stress at the surface of 308 MPa , which overcomes the tensile strength of basaltic glass ($\sim 10^8\text{ Pa}$; Webb and Dingwell, 1990). This confirms that *in situ* granulation by thermal stress is plausible at the margins of the lapilli during interaction with seawater. The vesicularity and vesicle size (e.g., Heap et al., 2014), and the permeability and crystallinity of the lapilli all influence the tensile strength of basaltic rocks and therefore affect *in situ* granulation by thermal stress. Thermal granulation can be a very fast process, as crack propagation velocity is expected to be in the range of hundreds to thousands of meters per second (e.g., van Otterloo et al., 2015). We do not rule out additional granulation induced by clast-to-clast collisions, but we believe it would be localized to isolated impact points

and is not sufficient to explain the observed textures. We therefore propose that thermal stress is the dominant cause of disruption.

Some particles generated by *in situ* granulation may have been spalled off, washed away, or dispersed by winds. Commonly, the observed jigsaw-fit ash particles are slightly rotated or displaced from their site of origin, intruding into the external vesicles of the scoria core and resulting in an ash rim with higher density than the core. Similar densification of jigsaw-fit particles was also observed in peperites and hyaloclastites (Hanson and Hargrove, 1999; Doyle, 2000). Inward displacement of some ash fragments into the external vesicles and tight packing of ash in the rims likely reflect a combination of (1) condensation of internal magmatic gas during quenching, causing suction and absorption of water and ash particles (cf. Allen et al., 2008), (2) compression of noncondensable magmatic gas such as SO₂ or CO₂ by the water column during cooling, and (3) particle collisions during transport. Vesicularity, vesicle connectivity, and permeability partly control the efficiency and depth of ash displacement into the particles during densification. Most of these mechanisms imply densification occurred either below the sea surface or shortly after ejection. After ejection, residual heat causes evaporation of brine and precipitation of salts (e.g., NaCl or MgSO₄), from both magmatic gases and seawater (Ayris et al., 2014), which may stabilize the “coated lapilli” (cf. Mueller et al., 2017). Our 3-D evidence from micro-XCT provides the first documentation of thermal *in situ* granulation for the microscale production of fine-ash particles in a volcanic eruption.

We propose a conceptual model to explain the formation of the ash-encased lapilli in three steps (Fig. 3): Following the initial magma fragmentation, there is (1) direct

contact between a primary pyroclast and seawater, causing thermal-stress–induced cracking and granulation. This is followed by (2) inward displacement of ash particles and seawater into the particle, causing densification of the ash rim and release of some outermost fragments generated by *in situ* granulation into the water column. Finally, (3) the ash-encased lapilli are injected into the atmosphere, possibly accompanied by precipitation of salts, which stabilize the ash rims and preserve the jigsaw-fit textures.

IMPLICATIONS FOR SUBAQUEOUS ERUPTIONS AND RELATED HAZARDS

Ash-encased lapilli are the dominant constituent of the lapilli fraction of deposits from the Surtsey (1963–1967; Moore, 1985), Capelinhos (1957–1958), and Hunga Tonga–Hunga Ha’apai (2014–2015) eruptions. These have always been termed “ash-coated” particles, with the inherent assumption of an active process where foreign ash particles are attracted to and adhere to the outside margins of a lapilli particle, i.e., similar to the aggregation process of ash into accretionary lapilli in moist atmospheric eruption plumes (Brown et al., 2012). The thermal cracks and jigsaw textures observed in all the lapilli examined in our study indicate that ash rims on scoriaceous lapilli may rather result dominantly from thermal granulation of particle margins. This implies a greater importance of this secondary disruption process than previously considered. Thermal granulation probably contributes to magma disruption during subaqueous eruptions occurring at any water depth and magma composition, producing particles with a broad range of sizes (e.g., the formation of **metric** pumices during the 2012 Havre eruption; Manga et al., 2018). In contrast to conventional dry and wet aggregation in volcanic ash plumes (Brown et al., 2012), the *in situ* granulation model binds at least some of the ash directly after generation. In this scenario, the ash-encased particles are actually an

indicator of ash production, rather than **sequestration** of free-ash particles into coatings. Additional wet aggregation of ash particles, or alternatively loss of ash from the rims, may occur during transport above sea level. Wet particle aggregation initiates at relative atmospheric humidity levels of 15%–20% or higher (Mueller et al., 2016), which is highly likely in Surtseyan eruption plumes. Interpreting ash-encased lapilli solely as the result of aggregation following primary fragmentation might cause an error in the inferred aggregation rate and total grain-size distribution, which are two essential eruption source parameters in models of tephra dispersal (e.g., Folch et al., 2010). In the future, understanding the conditions that alter the relative balance between in-plume aggregation (decreasing free ash) and the subaqueous production of ash by *in situ* granulation (possibly increasing free ash) will be a key for better assessment of potential hazard of ash particles in the atmosphere impacting **human** populations and air traffic.

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FIGURE CAPTIONS

Figure 1. Two- and three-dimensional (2-D and 3-D) textural properties of ash-encased

lapilli. Top, medium, and bottom rows correspond to samples CAP370–3–1, HH47–2,

and HH28–3, respectively. A–C: 2-D slices through X-ray computed microtomography

(XCT) data showing internal textures of lapilli. White line corresponds to boundary

between core and ash rim. Ash particles filling external vesicles of cores are common.

Note highly variable internal textures (vesicle size, elongation, size distribution, and

crystallinity of cores) in lapilli. D–F: 3-D XCT volume renderings illustrating cores

(blue) and presence of large cracks at their margins (red). Dashed circles in E serve to

highlight locations of smaller cracks in this sample. G: 2-D slice through XCT data of

sample CAP370–3–1 showing margins of core (pink outline) and associated ash rim with jigsaw-fit particles (outlined in blue solid lines). Smaller particles ($<20\text{ }\mu\text{m}$) could not be identified as jigsaw-fit due to voxel resolution. Particles 1 and 2 represent two fitting particles with vesicle concavity in common, and **their** fit with part of core (labeled 3 and visible as part of core in 3-D). H: Higher-magnification view of margin of sample HH47–2 showing jigsaw-fit texture in ash rim at margins of vesicular core (highlighted in pink solid line). Orange dashed line corresponds to vesicle concavity in common with several ash particles as well as with core. Note high number **and** density and small size of cracks in this area. I: Close-up of margins of sample HH28–3 showing cracks (red), ash rim, and fillings.

Figure 2. Jigsaw-fit textures. A–B: Backscattered-electron–scanning electron microscope images of sample HH37–3 showing jigsaw-fit textures at margin of dense core, with (B) four jigsaw-fit particles separated by cracks and sharing matching crystal fragments. C: X-ray computed microtomography (XCT) volume of jigsaw-fit particles 1 and 2 in Figure 1G viewed from two different angles (left and right). Matching vesicle concavities are highlighted in orange dashed circles. D–E: Two- (D) and three-dimensional (E) visualizations of XCT data showing vesicle with radial crack (red) distribution in samples HH47–2 and CAP372–2–3.

Figure 3. Conceptual model of formation of ash-encased lapilli. A: At time t_1 , a magmatic particle is ejected into water column and is in direct contact with water (in blue). High cooling rate induces a high thermal gradient at margins of particle;

subsequent quenching and high levels of thermal stress trigger cracking at margins of particle. Crack number and density **are** much higher at margins due to higher temperature contrast between particle and coolant, causing *in situ* granulation and formation of ash particles at outer parts of margins, showing jigsaw-fit textures. Thermal cracks are represented in red. B: At time t₂, inward displacement and rotation of ash particles toward core induce ash filling of external vesicles and densification of rim. Some outermost ash particles might also be spalled off after granulation and are released into water column. Arrows represent both spalling of some external particles released to plume and inward displacement of ash toward core during densification. C: At time t₃, once particle is well above water, residual heat in particle core leads to evaporation at margins and subsequent salt precipitation, enhancing stability of ash rims when deposited on land. Ash-encased particles can be easily identified with jigsaw-fit (highlighted in blue), whereas margins of the core are outlined in pink. Green rectangles represent salts binding ash particles in coating.

¹GSA Data Repository item 2019xxx, description of geological setting of study, description of methods for thermal stress analysis, and discussion on effect of crystals on crack propagation, and Table DR1 (scan conditions for ash-encased lapilli analyzed by XCT), is available online at <http://www.geosociety.org/datarepository/2019/>, or on request from editing@geosociety.org.